

~~J.E. Gordon~~

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THE APPLICATION OF THE GAUSSIAN PLUME  
EQUATION TO  $UF_6$  RELEASES

T. J. MAYO

K-25 INFORMATION  
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UNION  
CARBIDE

PADUCAH GASEOUS DIFFUSION PLANT  
PADUCAH, KENTUCKY

prepared for the U.S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION  
under U.S. GOVERNMENT Contract W-7405 eng 26

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THE APPLICATION OF THE GAUSSIAN PLUME  
EQUATION TO UF<sub>6</sub> RELEASES

T. J. Mayo

Technology Laboratory Department

UNION CARBIDE CORPORATION  
NUCLEAR DIVISION

Paducah Gaseous Diffusion Plant  
Paducah, Kentucky

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### A B S T R A C T

A series of  $\text{SF}_6$  releases has been made both inside one of the large gaseous diffusion plant buildings and outside of the plant restricted area to determine if the Gaussian plume equation can be used to predict atmospheric dispersion in the vicinity of the Paducah Gaseous Diffusion Plant. It was concluded that use of the equation would give reasonable, but probably conservative, estimates of downwind concentrations of contaminants as a result of gas releases in the diffusion plant.

A separate series of tests were made to examine the fallout of uranium and  $\text{HF}$ . Under relatively stable atmospheric conditions, it was concluded that some  $\text{UO}_2\text{F}_2$  fallout does occur, but it is believed that for distances of up to several hundred meters fallout would not be a major factor in reducing predicted downwind uranium concentrations.

## TABLE OF CONTENTS

INTRODUCTION . . . . .	5
SUMMARY . . . . .	6
ATMOSPHERIC DISPERSION TESTS . . . . .	6
Ground Level Releases . . . . .	7
Process Building Releases . . . . .	9
UO <sub>2</sub> F <sub>2</sub> FALLOUT TESTS. . . . .	13
PLUME DENSITY EFFECTS. . . . .	14
APPENDIX A . . . . .	16
APPENDIX B . . . . .	18

THE APPLICATION OF THE GAUSSIAN PLUME  
EQUATION TO UF<sub>6</sub> RELEASES

INTRODUCTION

The generalized Gaussian plume equation is the relation most frequently used to describe atmospheric dispersion of gases and aerosols. The equation for average ground level concentrations is:

$$\chi(x, y, \sigma) = \frac{Q}{\pi \sigma_y \sigma_z \bar{U}} \exp \left[ -\frac{1}{2} \left( \frac{y^2}{\sigma_y^2} + \frac{H^2}{\sigma_z^2} \right) \right]$$

where:

$\chi$  = the ground level atmospheric concentration in grams per cubic meter.

$Q$  = the source strength in grams per second.

$\bar{U}$  = the average wind speed in meters per second.

$\sigma_y, \sigma_z$  = dispersion coefficients in meters which are functions of atmospheric stability and downwind distance,  $x$ , along the plume axis.

$y$  = the crosswind distance in meters from the plume axis, assumed to be the same as the mean wind direction.

$H$  = the effective height of the release in meters and includes plume rise due to buoyancy and velocity.

Since it is intended to use this relation with the methods and dispersion coefficients given by Turner in the *Workbook of Atmospheric Dispersion Estimates*<sup>1</sup> to evaluate the consequences of the release of potentially hazardous gases at the Paducah Gaseous Diffusion Plant, applicability of this equation has been examined. These methods and dispersion coefficients are intended to describe dispersion over relatively open country or rural areas. The description is considered less reliable over urban areas due primarily to the influence of a city's larger surface roughness and heat island effects. A gaseous diffusion plant with its large structures and the discharge of considerable amounts of heat may, in effect, behave as a small city. Thus, there is a question concerning the applicability of the Gaussian plume equation and the dispersion coefficients given by Turner. This report summarizes the results of a series of tests conducted to investigate this question.

The Gaussian plume equation is applicable to aerosols and particulate matter smaller than about 20  $\mu\text{m}$ . The release of  $\text{UF}_6$  to the atmosphere, of course, rapidly results in a cloud of  $\text{UO}_2\text{F}_2$  particles and HF gas. The HF would be expected to disperse according to the equation unless it absorbs or reacts with its surroundings. The  $\text{UO}_2\text{F}_2$  should behave similarly if the particle size is in the proper range. Because these  $\text{UO}_2\text{F}_2$  clouds have not been well characterized, a series of  $\text{UF}_6$  releases was also made to investigate the fallout of the  $\text{UO}_2\text{F}_2$  and the loss of HF downwind of the release.

#### SUMMARY

In the vicinity of the Paducah plant, the Gaussian plume equation with the dispersion coefficients given by Turner<sup>1</sup> very satisfactorily describes atmospheric dispersion of ground level releases away from the influence of the plant microclimate. The microclimate influence is quite localized and normally would affect dispersion only in the immediate vicinity of the large buildings and cooling towers. For releases inside the plant where both the plant microclimate and plume rise may affect dispersion, results are more variable, but prediction by the Gaussian plume equation is still adequate and probably conservative in many cases.

It was concluded that during stable atmospheric conditions the fallout of uranium following a release is not rapid and, therefore, the effects of uranium cannot be ignored during such releases.

#### ATMOSPHERIC DISPERSION TESTS

In these tests the non-toxic gas sulfur hexafluoride ( $\text{SF}_6$ ) was used as a tracer. The use of this extremely inert gas eliminated complications due to deposition and simplified both the release and sampling phases of a test. The techniques employed in this work were investigated and developed by Saltzman, et al.,<sup>2</sup> of the Division of Air Pollution, Robert A. Taft, Sanitary Engineering Center, Cincinnati, Ohio.

Sampling was accomplished by means of battery-operated vacuum cleaner clothes brushes of the type available at department stores. A flow restrictor was used to give a sampling rate of about one liter per minute. Samples were taken in 30 cm by 60 cm. Saran bags with a wall thickness of 0.0025 cm. Analysis was by chromatographic means using an electron capture detector.

Since samples could not always be analyzed immediately, sample stability during the holding period prior to analysis was of utmost importance. This was investigated at two concentration levels. No detectable concentration change was seen at 3100 parts per trillion (ppt)  $\text{SF}_6$  in air in 72 hours or at 13 ppt in 120 hours. Saltzman investigated the effects of common atmospheric pollutants and found no problems.

Two series of tests were conducted. In the first series,  $\text{SF}_6$  was released outdoors at ground level directly to the atmosphere at locations away from the diffusion plant. The purpose of this series was to compare actual dispersion with the Gaussian plume predictions with the plant influences minimized. In the second series, the releases were made into a vertically discharging ventilation duct of a process building to introduce the factors of plume rise and plant microclimate.

In all tests the procedure was the same. The  $\text{SF}_6$  release rate was 54 g/min. The mean wind speed and direction were established and six to nine samplers set up roughly in a line to span the mean wind direction. Efforts were made to avoid heavily wooded areas, but the terrain was normally heavily vegetated with brush and different size trees. The sampling line was located 2000-3000 m downwind and the samplers were separated by 300-350 m. After the  $\text{SF}_6$  release was begun, time was allowed according to the mean wind speed to permit the  $\text{SF}_6$  to reach the sampling area. Then, four sets of samples were taken in rapid order. Each set was composed of one 10-minute sample taken simultaneously by each of the samplers.

During the runs, observations were made on cloud cover and solar altitude as well as wind speed. This information was used to determine the atmospheric stability category and to select the proper values for the dispersion coefficients. The stability categories were determined by the method suggested by Turner.<sup>3</sup>

Because of the normal meander of the wind during most of these tests, it was impossible to predict exactly which one of the downwind samplers would lie nearest the plume centerline. Nevertheless, it was necessary to assume that one of them would, and the method of comparison used was to accept the maximum concentration found in a set of samples as the maximum which occurred at the sampling distance and compare it with the expected value at that point calculated from the Gaussian plume equation. The assumption that the sampler was on the centerline would in some cases introduce a small bias since it is probable that the actual centerline was between two samplers. This error will be small because, at the distances involved, the crosswind dispersion is quite wide due to the meander.

#### Ground Level Releases

The conditions and results of the ground level release tests are summarized in table 1. The detailed test results are given in Appendix A, table A1. For the individual sample groups, the maximum concentrations found ranged from 36% to 191% of the value predicted by the Gaussian plume equation with  $y$  and  $H$  equal to zero. The overall average was 86%.



Table 1  
COMPARISON OF ACTUAL AND PREDICTED CONCENTRATIONS  
FOR GROUND LEVEL SF<sub>6</sub> RELEASES

<u>Run</u>	<u>Stability</u>	<u>Wind, m/sec.</u>	<u>Approximate Sampling Distance, m</u>	<u>Sample Group</u>	<u>Maximum Conc., ppt</u>	<u>% of Predicted Value</u>
5	B	2.2	2000	1	170	51
				2	320	96
				3	410	136
				4	360	108
6	B	1.8	2000	1	275	75
				2	355	92
				3	345	92
				4	310	68
7	C	3.3	2800	1	325	98
				2	120	36
				3	180	55
				4	130	39
8	B	2.5	2450	1	240	124
				2	240	129
				3	75	39
				4	355	191
10	C	3.0	2400	1	390	69
				2	230	41
				3	330	59
				4	660	117
Overall Average--						86

To provide a clearer overall picture of these tests, all the analytical results of run 6 are given in table 2. During this run, a shift in wind direction occurred as opposed to random meandering around a fixed mean direction as was the case in the other runs. This can be seen in the shift of the maximum concentration from sample point 6 in the first group through point 5 and to point 3 in the last group. This is an example of the problem of determining the mean wind direction for a distant ten-minute sample. It may also be noted that the distances to the sample points varied in run 6. This was typical of all runs since the samplers were positioned in easily accessible locations along roads and in cleared fields for convenience. This explains situations such as in table 1, run 6 where in group 1, 275 ppt is 75% of the predicted value while in group 4, 310 ppt is only 68%.

Table 2

## SUMMARY OF RUN 6

Sample Point:	<u>1</u>	<u>2</u>	<u>3</u>	<u>4</u>	<u>5</u>	<u>6</u>
Distance, m	1720	1640	1835	2130	2040	2065
<u>10 minute average concentration, ppt</u>						
Group 1	NF	5	50	85	180	275
" 2	NF	5	5	30	355	170
" 3	NF	NF	40	110	345	5
" 4	5	5	310	100	45	10

As events developed during these tests, there was reason occasionally to suspect that some results were too low. In run 5, group 1, for example, the overall results indicated that sampling was begun before the plume had fully reached the sampling line. In run 7 in all groups, the maximum concentration was found at the same sample point at one end of the sampling line which strongly suggests that higher concentrations occurred outside the sampling range. These two factors along with the distance between samplers would tend to produce results which were low compared with the expected maximum values, and overall the results were somewhat low. However, the Gaussian plume calculations are generally considered to be good to within a factor of three, and even considering the possibility of low results, all the values of table 1 appear to be within this range. There is, therefore, no reason to doubt the validity of the Gaussian plume calculations for this area under the conditions of these tests.

#### Process Building Releases

The second group of tests was conducted by releasing the SF<sub>6</sub> at cell floor level into the southernmost ventilation duct on the west side of C-333. This is one of several such ducts which exhaust air and waste heat from the cell floor and stage motors through the motor exhaust system. The ducts are 1.83 m by 11.0 m in cross section and discharge at roof level 25.3 m above the ground. The volume and temperature of the exhausted air depends primarily on the cascade power level and the outside air temperature.

The significant release and meteorological conditions which existed during these tests are summarized in table 3. Plume rise was calculated from Holland's relation<sup>4</sup> based on observation of steam plants in the vicinity of Oak Ridge.

Table 3

CONDITIONS DURING SF<sub>6</sub> RELEASES INTO  
C-333 VENTILATION DUCT

Run No.	<u>11</u>	<u>12</u>	<u>13</u>	<u>14</u>	<u>15</u>
Cascade power level, mw	1000	1500	1420	1440	1475
Effluent Air					
Temperature, °C	40	40	45	44	46
Velocity, m/sec.	7.0	7.0	6.0	3.3	7.3
Volume, m <sup>3</sup> /sec.	141	140	120	66	147
Atmospheric temperature, °C	25	18	14	4	18
Plume rise, m	18.1	20.9	15.9	12.5	14.6

The results of this series of tests are summarized in table 4. It can be seen from comparison with table 1 that these results are more scattered than were the results of the ground level releases. In terms of percent of the predicted value obtained from the Gaussian plume equation with y equal to zero, the results of runs 11, 12, and 13 lie outside the extremes of the ground level releases. In general, the values of runs 12 and 13 differ from the predicted values by a factor of about 4 or somewhat more than the factor of 3 which is expected.

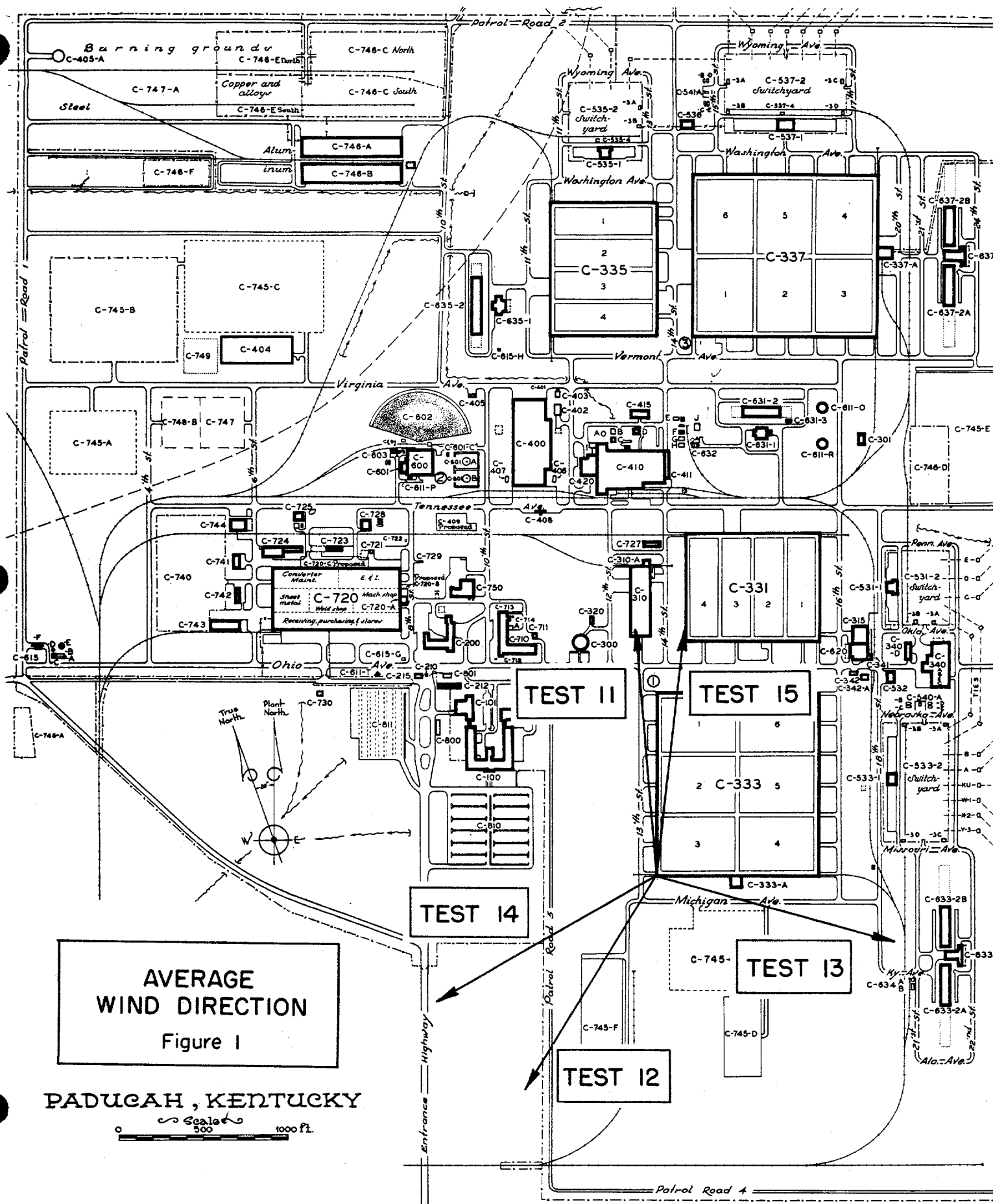
The run 11 values are extremely low; however, examination of all the data from this run, Appendix A, strongly indicates that the maximum plume concentrations lay outside the sampling range when group 1, 2, and 3 samples were being taken. The group 4 maximum suggests that run 11 might actually have been similar to run 13 in terms of percent of predicted value. In any case, the results are certainly less than the predicted levels.

In figure 1 the location of SF<sub>6</sub> release point and the average wind direction during the releases are shown. In runs 11 and 15, the wind direction was from the release point toward the major process buildings while in run 13 it was directed toward the C-633 cooling towers. In these three runs an average of only 53% of the expected SF<sub>6</sub> concentration was found. On the other hand in runs 12 and 14, in which the release would have been carried away from the plant, an average of 223% of the expected concentration was seen. Thus, it is indicated that there is a plant microclimate effect which generally provides

Table 4  
COMPARISON OF ACTUAL AND PREDICTED CONCENTRATIONS  
FOR VENTILATION DUCT SF<sub>6</sub> RELEASES

<u>Run</u>	<u>Stability</u>	<u>Wind, m/sec.</u>	<u>Approximate Sampling Distance, m</u>	<u>Sample Group</u>	<u>Maximum Conc., ppt</u>	<u>% of Predicted Value</u>
11	D	3.7	2300	1	120	11
				2	100	8
				3	35	3
				4	340	27
12	B	3.2	2800	1	310	410
				2	310	410
				3	260	260
				4	250	330
13	C	4.3	2700	1	110	37
				2	60	22
				3	60	24
				4	60	22
14	C	3.5	2700	1	530	106
				2	240	72
				3	480	177
				4	440	119
15	D	5.3	2400	1	1330	140
				2	830	87
				3	410	44
				4	670	71
Overall Average-						119

increased atmospheric dispersion. Considering both the limited data available and the variability of the data, no quantitative modification of the equation has been attempted. Rather, it is concluded that for releases inside the plant, concentration predictions based on the stability categories and dispersion coefficients given by Turner will be satisfactory. There is a considerable probability of the estimates being conservative depending upon the release location and wind direction, but no quantitative correction is believed possible because of the many possible influencing parameters.



UO<sub>2</sub>F<sub>2</sub> FALLOUT TESTS

With the applicability of the Gaussian plume equation established, one substantial question remains before reliable estimates of downwind uranium and fluoride concentrations can be made. Do the UO<sub>2</sub>F<sub>2</sub> and HF resulting from the hydrolysis of UF<sub>6</sub> remain airborne in the same manner as an inert gas? It is sometimes believed that during a UF<sub>6</sub> release most of the uranium will fall out of the atmosphere rapidly close to the release site and that HF will be the greatest health hazard offsite. A series of test releases of UF<sub>6</sub> was made to determine the composition of the UO<sub>2</sub>F<sub>2</sub> cloud downwind. The details of the tests are discussed in Appendix B and the results are summarized here.

Known mixtures of SF<sub>6</sub> and UF<sub>6</sub> were released outdoors and the generally visible cloud sampled at two distances as it dispersed downwind. The SF<sub>6</sub>, being entirely stable, acts as an internal standard. Uranium, fluoride (from UF<sub>6</sub>), and SF<sub>6</sub> were determined. It was intended to use the ratios of these materials found in the samples compared to the known ratios released to determine the loss of uranium and fluoride from the cloud. For example, if the U/SF<sub>6</sub> ratio in the released mixture was 10, but the ratio in the downwind samples was 5, this would indicate a loss or fallout of half of the uranium. For reasons which are not understood, however, the results were quite variable with the various ratios found frequently exceeding the known release values. While efforts were made to resolve this difficulty, it persisted which prevented attaining some of the original goals of the study. Nevertheless, some useful conclusions were reached. To magnify any fallout, all tests were made under stable weather conditions. From all the evidence, however, including the sampling results and visual observations of the release area and the clouds, it can be stated with some confidence that the losses of uranium were small, being almost certainly less than 20 to 25% in 400 m. There was also apparently some loss of fluoride, but to a lesser extent than uranium. Some fluoride would, of course, be lost as UO<sub>2</sub>F<sub>2</sub> when uranium settles out.

While the data from this investigation were not entirely unambiguous, it is believed that it cannot be assumed that most of the uranium will quickly fall out following a UF<sub>6</sub> release. This conclusion, of course, applies to outside releases where no attempt is made to contain any of the material or to reduce the concentrations by water sprays or the like. It would apply, however, to any part of a release escaping from a building regardless of whether or not a major portion of the original release was captured by whatever means.

The UO<sub>2</sub>F<sub>2</sub> particles are predominantly in the range of 1 to 20  $\mu$ m, and contrary to some assumptions, the HF formed in the hydrolysis appears to be largely present in the atmosphere as free HF.

## PLUME DENSITY EFFECTS

It is known that plumes significantly more dense than the surrounding air will exhibit descending rather than ascending motion. Bodurtha, et al<sup>5</sup>, have examined this, and one of their conclusions is that the usual atmospheric dispersion equations cannot be used with reliability to estimate the concentration of dense stack gases at the ground. They emphasize, however, that they are not talking about gases containing a small fraction of high molecular weight material, but of gases with a net molecular weight of the total stream significantly greater than the molecular weight of air (29). This would apply also for density differences due to temperature effects, of course.

While  $\text{UF}_6$  is a high molecular weight gas (352), this fact does not prohibit the use of the Gaussian plume relations for our purpose which is to estimate ground level concentrations 1.5 miles and farther from the release source. We are interested in two different situations. The first is a ground level  $\text{UF}_6$  release which could occur outdoors. The second is a release in which  $\text{UF}_6$  would escape initially indoors.  $\text{UF}_6$  would then be expelled to the atmosphere from an elevated location in a greatly diluted form.

Consider an outdoor-ground level  $\text{UF}_6$  release of 757 g/s (100 lb/min) during the stable F atmospheric condition with a very slight breeze of 1 m/s (2 mph). Assume for purposes of illustration that the  $\text{UF}_6$  remains unhydrolyzed. This is, of course, a condition of minimum dispersion during which plume density effects would be the greatest. Since the release is at ground level, the plume obviously cannot descend, but the initial high density would undoubtedly reduce dispersion in the vertical direction somewhat.

From the graphs of dispersion coefficients<sup>1</sup>, it will be seen that at a distance of 200 m from the source the horizontal and vertical coefficients are 7.6 and 4 m, respectively. These coefficients are standard deviations of the concentration distributions within the plume; therefore, essentially all plume material would be contained within  $\pm 2$  standard deviations of the plume centerline. The plume cross section at 200 m, calculated as half an ellipse with semiaxes equal to  $2\sigma_y$  and  $2\sigma_z$ , would be 191 m<sup>2</sup>. From this value, the wind speed and the  $\text{UF}_6$  release rate, the average plume concentration 200 m from the source is calculated to be about 250 ppb on a volume basis. This corresponds to a density only 1.003 times that of air. This is, of course, an estimation, but it does indicate that even under very unfavorable conditions the early dispersion is rapid enough to eliminate plume density effects quickly.

Any reduced dispersion near the source would have a very minor effect on concentrations 2400 m downwind. For example, assume no dispersion at all in the first 200 m which is the same as moving the release point 200 m downwind. This would increase the estimated concentration by only 15% in the example above.

In the case of a release in a process building, the  $\text{UF}_6$  would mix with the building air; then be ejected by the building ventilation system in a highly diluted form. For example, at maximum ventilation each of the motor exhaust stacks on a 000 building would discharge about  $5500 \text{ m}^3$  of air per minute. A  $\text{UF}_6$  release of  $100 \text{ lb/min}$  would correspond to roughly  $3 \text{ m}^3/\text{min}$ . Should all the  $\text{UF}_6$  be discharged from one stack, the concentration would be about 500 ppm on a volume basis. The average density would be approximately 1.006 times that of air. Further dilution would quickly follow escape from the building. As in the previous case, the effect of plume density on concentrations 1.5 miles from the source would be insignificant.

#### REFERENCES

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- <sup>2</sup> Saltzman, B. E., et al, *Analytical Chemistry* 38, 6, p. 753 (1966).
- <sup>3</sup> Turner, D. B., *Journal of Applied Meteorology*, February, 1964.
- <sup>4</sup> Briggs, G. A., *Plume Rise*, TID-25075, p. 22 (1969).
- <sup>5</sup> Bodurtha, F. T., et al, *Chemical Engineering Progress*, Vol. 69, No. 4, April 1973.







## APPENDIX B

When  $\text{UF}_6$  is released to the atmosphere, it is quickly hydrolyzed to  $\text{UO}_2\text{F}_2$  and HF by atmospheric moisture. It is frequently believed that a large part of the  $\text{UO}_2\text{F}_2$  rapidly falls to the ground due to gravitational effects while the HF remains airborne. On the other hand, in atmospheric dispersion work it is generally assumed that aerosols (particle diameters less than 20  $\mu\text{m}$ ) remain airborne for long periods of time. The gaseous reaction of  $\text{UF}_6$  and  $\text{H}_2\text{O}$  frequently produces  $\text{UO}_2\text{F}_2$  particles of micron or submicron size or, in other words, well down in the aerosol range. There is, therefore, some question about the rapid fallout of  $\text{UO}_2\text{F}_2$ . There is also a possibility that HF, with its strong tendency to adsorb on practically any surface, would attach to the  $\text{UO}_2\text{F}_2$  particles and also be removed from the air should solids fallout occur.

The method chosen to examine the possible loss of uranium and HF from the atmosphere was to release known quantities of gaseous  $\text{UF}_6$  and  $\text{SF}_6$ , sample the atmosphere downwind, and determine the total U,  $\text{F}^-$ , and  $\text{SF}_6$  in the sample. Losses of either U or  $\text{F}^-$  should be reflected in deviations of the ratios of U and  $\text{F}^-$  to  $\text{SF}_6$  from the known ratios which were released. Loss of either U or  $\text{F}^-$  relative to the other would also be reflected, of course, in the U to  $\text{F}^-$  ratio.

Approximately 215 g of  $\text{UF}_6$  and 14 g of  $\text{SF}_6$  were charged into a 5.6-liter Monel bulb with the amounts established to within 1-2%. The bulb was then heated to 230°F to vaporize the  $\text{UF}_6$ . At this temperature, the vapor pressure of  $\text{UF}_6$  is about 76 psia. In the bulb employed, the actual  $\text{UF}_6$  pressure was about 50 psia at 230°F with an additional 8 psi supplied by the  $\text{SF}_6$ . The hot bulb was then carried into the field in an insulated box (also heated to 230°F) and the  $\text{UF}_6$ - $\text{SF}_6$  gaseous mixture released. The mixture pressure was monitored to make certain that there was no  $\text{UF}_6$  condensation before release. About 75% of the bulb contents, or approximately 160 g of  $\text{UF}_6$  and 10 g of  $\text{SF}_6$ , were released during each run. The releases were essentially complete in less than half a minute.

Sampling was done with four portable Bendix Model 15003 battery-operated air samplers. The air was pulled through two  $\text{K}_2\text{CO}_3$ -coated, membrane-type filters to trap the  $\text{UO}_2\text{F}_2$  and HF. The sampler discharge was then split and a known fraction (about 14%) trapped in a vinyl bag for  $\text{SF}_6$  determination. Occasionally an uncoated filter was used in front of the two coated filters to trap particles and determine the amount of adsorbed HF.

The U and  $\text{F}^-$  analysis were by standard wet chemical methods. The  $\text{SF}_6$  analyses were by chromatographic means using an electron capture detector.

The releases were made on the plant site in cleared areas generally over mowed fields. In order to obtain stable conditions, it was necessary to make the releases in the last half hour before sunset. At this time of day when the sky is clear and the wind speed low, the atmosphere becomes stable in the lower levels as an inversion builds up. Gravitational effects on  $\text{UO}_2\text{F}_2$  should be greatest at this time.

Each cloud was sampled at two different distances. At each distance, two samples were taken by positioning the samplers directly in front of the approaching cloud at the last moment. The results are summarized in table B1. The values for each distance are the averages of the two samples mentioned above. It was not possible to calculate meaningful expected concentrations since the releases were very brief and actual sampling time was short, 4-6 minutes.

Table B1

SUMMARY OF  $\text{UF}_6$ - $\text{SF}_6$  RELEASE RESULTS

Run	Sampling Distance, Meters	Ratios		
		U/F	U/ $\text{SF}_6$	F/ $\text{SF}_6$
7	90	2.6	10	3.8
	210	2.1	21	9.9
8	190	2.8	17	6.3
	380	2.3	12	5.2
9	240	2.7	15	4.9
	480	-	10	-
10	70	2.1	29	14
	440	1.6	17	10
11	70	1.5	12	7.9
	440	2.0	11	5.4
12	70	2.1	14	6.5
	440	1.9	9	4.7
Theory		2.1	10	4.7

Examination of table B1 shows that while the ratios generally decrease with distance there are cases, runs 7 and 11, where the reverse is true. It will also be noted that more often than not the ratios ex-

ceed the known release values. While there are possible explanations for such results, they are generally not persuasive so it was believed that additional runs would do little to clarify the situation.

There are, however, some worthwhile observations which can be made from the data. Because of the variability of the data, probably the best way to look at it is from the standpoint of overall averages which are summarized in table B2. Considering the ratios in the order presented, the U/F ratio change from the near to the far sample point indicates a small loss (13%) of uranium relative to the fluoride. The fluoride here is defined as the fluoride in the  $\text{UO}_2\text{F}_2$  plus HF from the hydrolysis. A similar loss of uranium (18%) is indicated by comparison to the internal standard  $\text{SF}_6$ . Only a minor loss (3%) of fluoride is indicated. At best, these values can only be considered as semiquantitative estimates because of the obviously poor precision of the data. It is concluded that some loss of uranium does occur downwind from an outside release during relatively stable weather conditions. The extent of the loss is not well defined, but it is probably small and almost certainly does not exceed 20-25% in 400 m. Presumably larger particles would fall out early, therefore, gravitational losses would be expected to be much slower at greater distances.

Table B2

## SUMMARY OF AVERAGE RESULTS

<u>Sampling Location</u>	<u>U/F</u>	<u>U/SF<sub>6</sub></u>	<u>F/SF<sub>6</sub></u>
Near	2.3	16	7.2
Far	2.0	13	7.0
% Decrease	13	18	3

While all releases were made during what was hoped to be very stable weather conditions, Pasquill's F category, it was apparent from the cloud behavior that this was not always the case. Conditions during run 7 were very stable, and the cloud stayed near the ground permitting extended observation. This observation of the cloud over 800-1000 m did not suggest a large loss of material. In the other runs, the atmosphere was not as stable and the cloud dispersed somewhat more rapidly. In neither condition was fallout seen to be a major factor, and the data support this conclusion.

To investigate the possibility of HF being adsorbed on  $\text{UO}_2\text{F}_2$  or atmospheric dust, in five runs, one of the samplers was provided with an untreated filter in front of the  $\text{K}_2\text{CO}_3$ -treated filters. This untreated

filter would catch the  $\text{UO}_2\text{F}_2$  and other dust while free HF would pass through and be caught by the  $\text{K}_2\text{CO}_3$  on the following filters. In the five runs, the portion of the total fluoride found on the untreated filter ranged from 36 to 50% with the average being 43%. Since 33% of the fluoride would be present as  $\text{UO}_2\text{F}_2$ , this indicates that only about 15% of the free HF was adsorbed on material trapped by the first filter. The fluoride considered here is only that from  $\text{UF}_6$  since  $\text{SF}_6$  does not hydrolyze.

In the above multiple filter experiments, the treated and untreated filters were also analyzed separately for uranium. Although the quantity of uranium found on the untreated filters ranged up to 500  $\mu\text{g}$ , none was detected on the treated filters. The lower limit of detection was 5  $\mu\text{g}$ . Since the hole size of the untreated filters was 0.8  $\mu\text{m}$ , it is apparent that submicron particles represented an insignificant portion of the total uranium released. Thus, it is indicated that the majority of the  $\text{UO}_2\text{F}_2$  is in the range of 1 to 20  $\mu\text{m}$ . Presumably, different hydrolysis conditions might produce other particle sizes.

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